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EXHALATION RATES DETERMINED IN CONSTRUCTION MATERIAL PRODUCED FROM NIOBIUM WASTE

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ABSTRACT

Mining operations annually generate large volumes of mining waste that can have several environmental impacts requiring appropriate management strategies in the short- and long-term. Recently, in Brazil, we have witnessed the catastrophic consequences of poor management of these tailings. One way to better manage mining waste may be to reduce its quantity by reusing it. In the case of niobium industry one possibility for the huge amount of solid tailing generated is its application as construction material. Nevertheless, these tailing also carries some radioactive elements. Exposure to radiation originated from construction material is mainly due to the presence of radionuclides from uranium and thorium series. The external exposure comes from the gamma emitters present in the material while internal exposure can come from radon and thoron gas. This study had evaluated the radiological impact of using niobium waste tailing in concrete plates and proof bodies, for use in construction material. It was determined the activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Th, ^{228}Ra and ^{40}K in cement made with niobium waste to compare with commercial cement. Exhalation rates for ^{222}Rn and ^{220}Rn were determined for cement plates and proof bodies made with 0, 50 and 100% of niobium waste. Results showed that the exhalation rates varied from 0.032 ± 0.004 to 0.20 ± 0.01 Bq/m^2.s for ^{222}Rn and from 0.044 ± 0.005 to 0.8 ± 0.1 Bq/m^2.s for ^{220}Rn in the different cements. The exhalation rate of the plates varied from 0.035 ± 0.002 to 0.35 ± 0.02 Bq/m^2.s for ^{222}Rn and from 0.048 ± 0.006 to 1.2 ± 0.1 Bq/m^2.s for ^{220}Rn and the variation for the proof bodies was 0.0042 ± 0.0006 to 0.2 ± 0.1 Bq/m^2.s and from 0.008 ± 0.001 to 0.054 ± 0.006 Bq/m^2.s for ^{222}Rn and ^{220}Rn, respectively.

1. INTRODUCTION

Radionuclides present in materials of natural origin, such as minerals and ores, or in those released in industrial process may pose a risk to workers, members of the public or the environment. They are commonly known as NORM – naturally occurring radioactive material. Mining and other industrial activities that involve minerals and raw material have

the potential to increase the effective dose received by individuals from natural sources [1]. During the extraction of minerals from the Earth's crust and their physical or chemical processing, the radionuclides may become unevenly distributed between the various materials arising from the process [2].

Mining operations annually generate large volumes of mining waste that can have several environmental impacts requiring appropriate management strategies in the short- and long-term. Mining waste are part of the materials resulting from the exploration and processing of ores consist of non-mineralized and low-grade mineralized orebodies [3]. After the treatment of minerals by separation processes solid waste or slurries may remain caring variated amounts of trace elements, including radionuclides.

Recently, in Brazil, we have witnessed the catastrophic consequences of poor management of these tailings. One way to better manage mining waste may be to reduce its quantity by reusing it. This concept has played a crucial role in maximizing the utilization of resources aiming to reduce environmental impacts [4]. A key focus on attaining sustainability in mining activities is an effective waste removal within the production processes.

One of the main applications of mining wastes is its use as construction material [5 - 9] mainly in cement and concrete industry [10, 11]. Nevertheless, the use of mining waste as recycled materials, demand safety requirements for its application due to the possibility of the presence of toxic compounds and the exposure from naturally occurring radionuclides [12].

Radiation exposure to the whole body occurs mainly from external gamma rays of ^{238}U, ^{232}Th decay series and, ^{40}K. On the other hand, internal exposure occurs by inhalation of radon that emanates from soil and rock in the ground, as well as building materials. There are three radon isotopes: ^{222}Rn (radon), ^{220}Rn (thoron), and ^{219}Rn (actinon) which half-lives are 3.82 d, 55.6 s and less than 4 s, respectively. Due to their very short half-life, thoron and actinon contribution to the total dose received, even in closed environments, are generally not taken in account, unless in specific cases [13].

The radiation exposure may increase for those living in houses built from materials with radiation levels above normal background [14], as it may occur when using recycled materials from mining process. Besides, outdoor and indoor temperature, air pressure, humidity fluctuations, porosity, pore distribution and pore type, type of surface treatment at the building site, and type of the coating material applied, may also influence radon exhalation in dwellings.

The possibility of increase in radon concentration inside dwellings, due to the use of mining waste as construction material, makes necessary the radiological assessment of the material. Hence, the concentration of ^{238}U and ^{232}Th and their decay products, as well as the exhalation rates for the raw material and in the construction material obtained should be determined. In this study it was evaluated the radiological impact due to radon inhalation caused by the use of niobium waste tailing in concrete plates and proof bodies for construction material. The activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Th, ^{228}Ra and ^{40}K was determined in concrete made with niobium waste to compare with commercial aggregate and cement. Exhalation rates for ^{222}Rn and ^{220}Rn were determined for concrete plates and proof bodies made with 0, 50 and 100% of niobium waste.

2. METODOLOGY

2.1. Samples description

The samples analyzed in this study were taken from a niobium mine located at Catalão city, in the state of Goias, Brazil, named CMOC International Brasil. The following samples were analyzed: one sample of sand, crushed stone, gravel 0 and gravel 1 from CMOC, named here as aggregates, and two other commercial aggregates of the same sizes from other regions near Catalão; one sample of the tailing without sizing; one sample of cement commercialized in the regular market; concrete plates and proof bodies were also manufactured with the proportion of 0, 50 and 100% of the waste material.

2.2 Gamma spectrometry

Gamma spectrometry was used to measure the activity concentrations for ^{226}Ra, ^{210}Pb, ^{228}Th, ^{228}Ra, and ^{40}K in samples of cement, tailing, sand, crushed stone, gravel 0 and gravel 1, and powdered proof bodies. For these measurements, the samples were sealed in polypropylene plastic cans (50 ml), in order to avoid radon loss, for at least 30 days before the measurement, to ensure equilibrium between ^{226}Ra and its short half-life progeny [15]. Activity concentrations were determined using a HPGe detector, model GX 2020, from Canberra Industries with 1.50 keV and 2.5 keV of resolution for the 122 keV and 1332 keV of ^{57}Co and ^{60}Co, respectively. The reference materials RGU, RGTh and RGK, from IAEA, were used for efficiency calibration and calibrated sources of ^{60}Co, ^{137}Cs, ^{152}Eu and ^{241}Am, were used for energy calibration. For the determination of ^{226}Ra activity concentration, the mean value of three decay transitions of its progeny, 295 keV and 352 keV of ^{214}Pb and 609 keV of ^{214}Bi, were taken. The Lower Limit of Detection (LLD) for this measurement configuration was 1.8 Bq kg^{-1}. For ^{228}Ra activity concentration determination, the mean values of the decay transition in 338 keV and 911 keV of ^{228}Ac were used (LLD of 3.6 Bq kg^{-1}), and the mean values of the decay transition in 238 keV of ^{212}Pb and 727 keV of ^{212}Bi were used for the determination of ^{228}Th (LLD of 4.0 Bq kg^{-1}). The decay transition in 1460 keV energy was used to directly determine ^{40}K activity concentration (LLD of 17 Bq/kg).

2.3 Neutron activation analysis

Determinations of ^{238}U and ^{232}Th activity concentrations were carried out by neutron activation analysis [16] in the same samples submitted to gamma spectrometry. For this measurement, approximately 150 mg of each sample was weighed and packed in plastic polyethylene bags, pre cleaned with a diluted solution of nitric acid. Samples were irradiated together with two reference materials (RM), USGS STM-2 and NIST SRM 1646a, and a paper filter was pipetted with a standard solution of the elements of interest. The sample concentration was calculated for each sample in relation to each reference material and the final report of the results is the mean value related to each RM. Samples and RMs were irradiated for 8 h in the IEA-R1 research reactor, at IPEN under a thermal neutron flux of 10^{12} cm^{-2} s^{-1}. After a decay time of 7 days the induced activity of uranium was determined and the one for thorium, after 15 days.

The counting time was 1 h was used for determination of the activity concentration of each sample and RM. Gamma spectrometry was performed by using an EG&G Ortec HP-Ge Gamma Spectrometer detector (AMETEK Inc., USA) and associated electronics, with a resolution of 0.88 and 1.90 keV for ^{57}Co (122 keV) and ^{60}Co (1332 keV), respectively. The analysis of the data was carried out by using in-house gamma ray software (the VISPECT program) to identify the gamma-ray peaks. The methodology evaluation was performed by cross-checking the reference materials and synthetic standards.

2.4 Radon and thoron measurement

For ^{222}Rn and ^{220}Rn, RAD7 (manufactured by Durridge Co., Inc.) detector was used. RAD7 is an alpha particle detector that converts alpha radiation into an electric signal, being able to distinguish the energy of each particle, which allows the identification of the isotopes (^{218}Po and ^{214}Po from ^{222}Rn and ^{216}Po from ^{220}Rn) produced by radon decay. RAD7 calibration is done at Durridge's indoor facility, using a radon chamber with a radium (^{226}Ra) source to provide a known flux of ^{222}Rn. According to the RAD7 User Manual the reproducibility is generally better than 2% and calibration accuracy is in the range of 5% [17].
Exhalation rate was calculated following the formula given equation 1 [18].

$$E_0 = \frac{Cx\lambda_{eff}xV}{1-e^{-\lambda effxt}} \quad (1)$$

Where, C is the equilibrium concentration, in Bq m^{-3}; λ_{eff} is the effective decay constant (s^{-1}), valid if leakage of radon and thoron out of the container is possible and if the activity concentration in the container air is low compared to the activity concentration in the pore air of sample; V is the effective volume of the chamber containing the sample plus tubings and the measurement chamber of the radon monitor, in m^{-3}; and t is time (s). The approach used in this study followed De Martino et al. [18], applying a non-linear curve fit to experimental data C versus t, in the form $y = a (1 - e^{-bx})$, where $a = E_0/ \lambda_{eff} \times V$ and $b = \lambda_{eff}$. The exhalation rate determined by the non-linear exponential regression is exemplified in Figure 1.

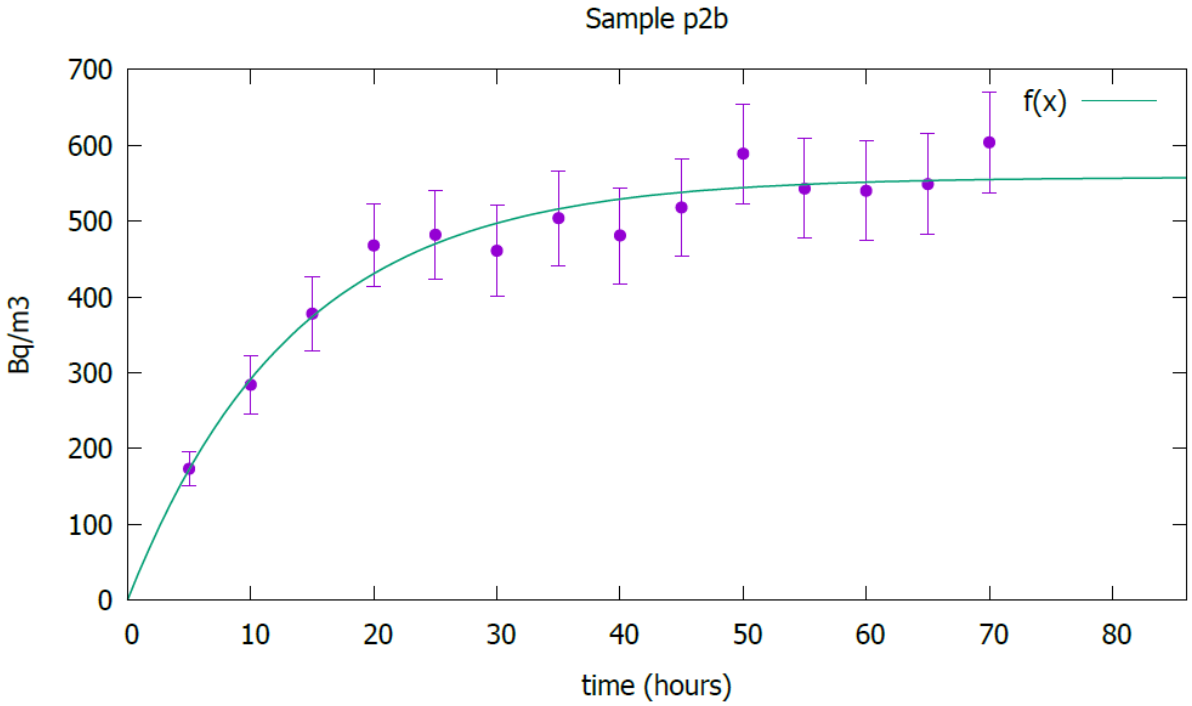

Figure 1: Experimental arrangement for radon exhalation rate measurement in powdered samples (a), plates (b), and proof bodies (c).

All measurements were done in a closed system. For exhalation rate determination of the powdered samples, an aliquot of approximately 300 g of each sample was placed inside an acrylic cylinder with 5 cm diameter and 200 cm length. For the plates, each one was covered with a plastic film 2 mm thick and sealed at the edges with adhesive tape to prevent Rn from escaping. The proof bodies were put inside a plastic can, also sealed with adhesive tape. The experimental arrangement for these measurements is shown in Figures 2, 3, and 4.

Figure 2: Experimental arrangement for radon exhalation rate measurement in aggregate samples.

Figure 3: Experimental arrangement for radon exhalation rate measurement in plate samples.

Figure 4: Experimental arrangement for radon exhalation rate measurement in proof bodies samples.

3. RESULTS AND DISCUSSION

The activity concentrations measured in the tailing from CMOC with no sizing applied (tailing-NB), cement acquired in the local market (cement), sand (S), crushed stone (CS), gravel 0 (G0), and gravel 1 (G1) from CMOC (NB) and from also from local market (G and PB) are shown in Table 1. For all samples, the activity concentration of the radionuclides from U series is relatively low, lower than 125 Bq kg^{-1}, and only ^{210}Pb presents higher concentrations in gravel 0 and 1 in the CMOC samples. Eštoková and Palaščáková [20] reported a compilation of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations in cements found in 16 different countries whose values varies from 12 to 92 Bq kg^{-1}; 14 to 59 Bq kg^{-1} and 93 to 1133 Bq kg^{-1}, respectively. Özdis et al. [21] compiled information given in 19 articles from 14 countries for the activity concentrations of the same radionuclides having found values in the range of 20 to 119 Bq kg^{-1} for ^{226}Ra, from 9 to 59 Bq kg^{-1} for ^{232}Th and from 51 to 2493 Bq kg^{-1} to ^{40}K. For the nuclides of U series and ^{40}K, both cements are in the same range of concentration. The same occurs for the nuclides of Th series for the marketed cement obtained in Catalão. The activity concentration of Th series nuclides in the concrete made

with CMOC waste, on the other hand, presented much higher values. The same can also be observed for the sand, crushed stones, gravel 0 and gravel 1 from CMOC. This amount of thorium is related to the mineralogy of the mined material composed by phyllite, carbonatites and amphibolites [22], also associated with high rare earth elements concentration.

Table 1: Activity concentrations measured in the cement, tailing (T), sand (S), crushed stone (CS), gravel 0 (G0) and gravel 1 (G1) samples from CMOC (NB), and commercialized in the region

Sample	^{238}U		^{226}Ra		^{210}Pb		^{232}Th		^{228}Th		^{228}Ra		^{40}K	
	Bq kg^{-1}													
Cement	19	±2	23	±1	7	±11	21	±1	28	±5	24	±4	247	±5
T-NB	ND		76	±3	ND		4839	±299	2269	±18	2162	±13	182	±10
S-NB	16	±9	65	±2	66	±16	1050	±54	854	±8	800	±7	853	±8
CS-NB	71	±11	57	±3	645	±14	1651	±102	820	±11	725	±9	947	±11
G0-NB	89	±6	42	±2	789	±13	1529	±86	902	±15	844	±6	869	±9
G1-NB	81	±6	57	±3	ND		1249	±70	738	±9	689	±6	834	±10
S-G	121	±8	113	±2	359	±5	122	±8	71	±7	65	±4	627	±3
CS-G	125	±4	77	±2	118	±15	142	±7	58	±11	62	±7	650	±14
G0-G	109	±3	43	±15	104	±5	102	±5	61	±7	56	±2	637	±8
G1-G	105	±3	94	±2	132	±5	110	±6	67	±3	58	±4	659	±9
S-PB	107	±4	59	±2	96	±10	210	±12	107	±7	98	±4	624	±8
CS-PB	121	±5	55	±2	99	±6	1192	±67	100	±8	95	±4	646	±9
G0-PB	95	±4	50	±3	ND		214	±11	92	±4	93	±4	580	±8
G1-PB	87	±5	59	±2	ND		159	±10	112	±8	105	±4	575	±8

Figures 5, 6, and 7 shows the distribution of the radionuclides from the U and Th series and ^{40}K in the powdered proof bodies manufactured with 0, 50% and 100% of CMOC material. For the elements of the U series (Figure 5), ^{238}U shows relatively low variation as well ^{226}Ra. Lead-210 is the only nuclide that presents a wide range varying from 24 to 190 Bq kg^{-1}. This can be due to the great variation of lithologies found in the mining place resulting in the gravel used to make these proof bodies.

For the elements of the Th series (Figure 6), it is observed that the activity concentrations for ^{232}Th, ^{228}Th and ^{228}Ra follows the pattern of the amount of CMOC material employed in the manufacture of the concrete proof bodies. Is also seems that the use of 50% of the waste reduces significantly the activity concentration of these nuclides in the final product. For ^{40}K, as for U nuclides series, only relatively small variation occurs (Figure 7).

Data on the radiological hazard indices: Radium equivalent, external hazard index, internal hazard index and external gamma absorbed dose rate for sand, crushed stone, gravel 0 and gravel 1 samples presented in this study are published in El Hajj et al. [23].

Concrete generally takes from 12–16% of cement, by weight, and also requires from 84–88% of coarse and fine aggregates mined from either hard rock sources, and then crushed, or from sands and gravels [24]. Using recycled material from mining operations may both contribute to the reduction of waste generated by mining processes as well as reduce the rock mining for cement and concrete industry itself, provided the radiological safety of the used material.

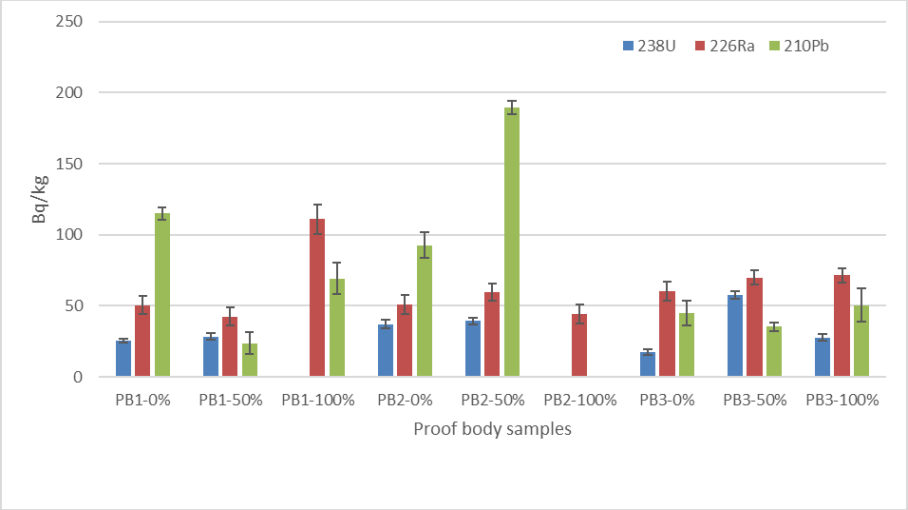

Figure 5: Activity concentration for the elements of U series in the samples proof bodies samples made with 0, 50 and 100% of CMOC material.

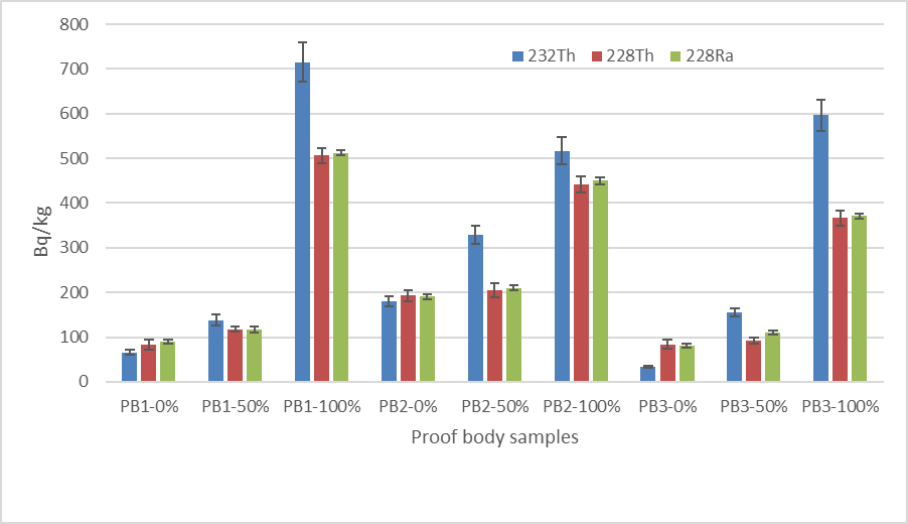

Figure 6: Activity concentration for the elements of the Th series in the proof bodies made with 0, 50 and 100% of CMOC material.

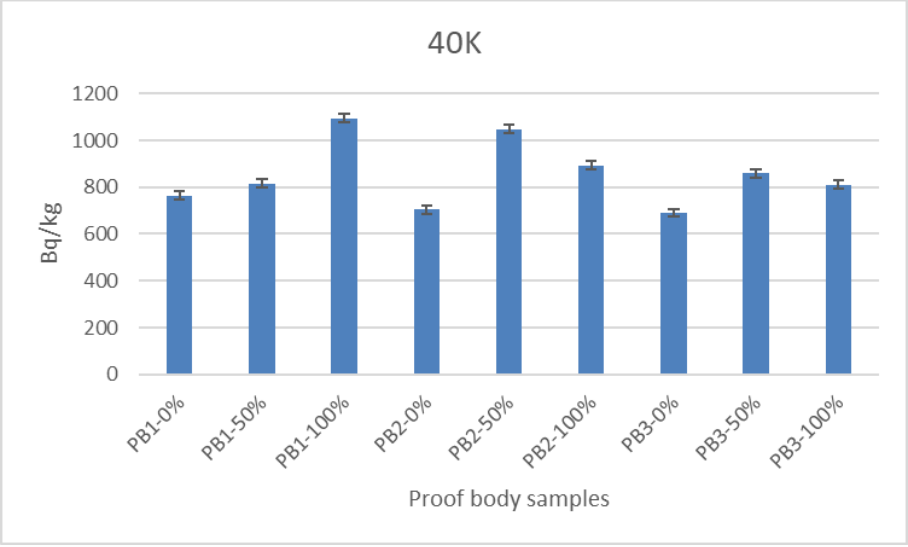

Figure 7: Activity concentration for ^{40}K in the samples of proof bodies made with 0, 50 and 100% of CMOC material.

3.1. Radon and thoron exhalation rates

The results for radon and thoron exhalation rates (ER) for the aggregates are presented in Figures 8. The aggregates samples showed a higher exhalation rate than the aggregates found in the region, both for ^{222}Rn and ^{220}Rn. Besides, the ER for thoron was three orders of magnitude higher than for radon, as it is normally found for different material applied for construction showed in Table 2.

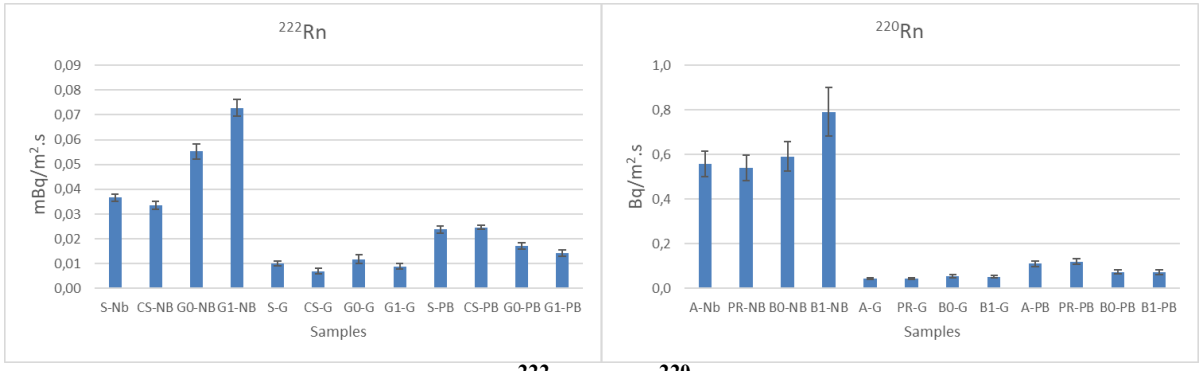

Figure 8: Exhalation rate of ^{222}Rn and ^{220}Rn in the aggregate samples.

The ER obtained for the plate samples , presented in Figure 9, showed that the plates made with cement type CPII (P1, P2, P3) show higher exhalation rates, both, for ^{222}Rn and ^{220}Rn than plates made with cement CV-ARI type (P4, P5, P6). This may indicate that the last cement can provide a less porous structure for the concrete reducing the radon and thoron scape.

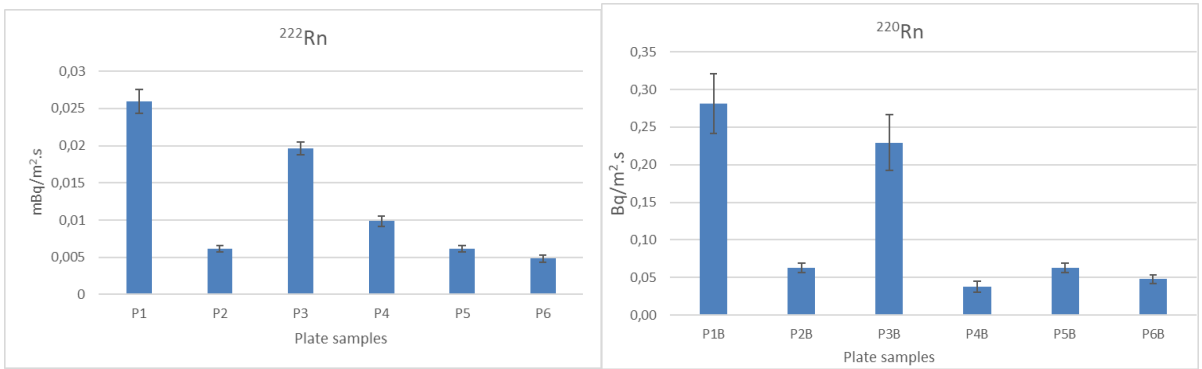

Figure 9: Exhalation rate of ^{222}Rn and ^{220}Rn in the plate samples.

The results obtained for the radon and thoron ER of the proof bodies are shown in Figure 10. The result for sample CP1-50% could not be determined for radon, and a high value with high uncertainty was observed for sample CP2-0%. Radon ER was also much lower than that observed for thoron, but no clear correlation could be derived from these data. One possibility that should be investigated is the bulk density and total pore volume of each proof body.

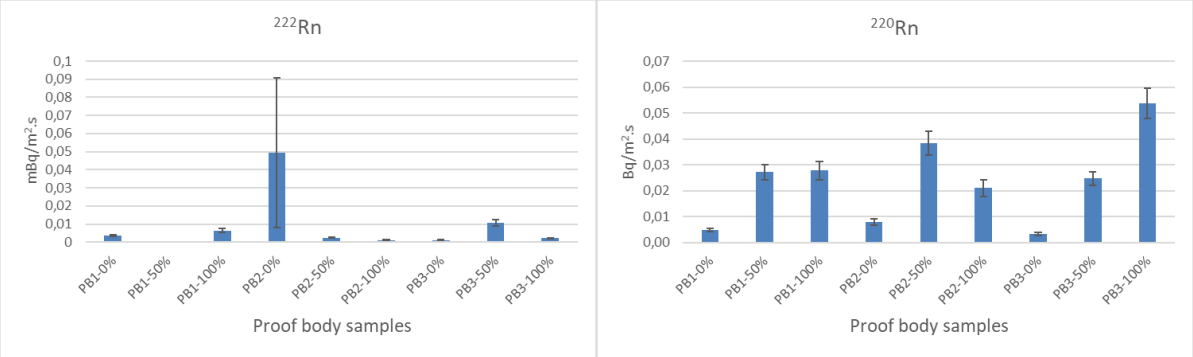

Figure 10: Exhalation rate of ^{222}Rn and ^{220}Rn in the proof bodies samples.

4. CONCLUSIONS

Aiming to test the feasibility of using aggregates from the niobium mining as construction material, this study evaluated the exhalation rate for ^{222}Rn and ^{220}Rn in aggregates (sand, crushed stones, gravel 0 and gravel 1), plates made with concrete and proof bodies produced using those aggregates. Additionally, the activity concentration of the U and Th series nuclides and ^{40}K were also evaluated.

The activity concentrations of Th series nuclides are much higher in CMOC aggregates samples than observed for the same type of material obtained in local market. These high activities justify the need of thoron measurements, despite its low half-life. The use of this material in the proportion of 50% may reduce the total activity concentrations of the final product. The exhalation rates for ^{222}Rn and ^{220}Rn in the plates and proof bodied produced with the aggregates, resulting from the niobium mining operations, are in the same range of common materials regularly employed in construction, and the use of cement of CV-ARI type resulted in a concrete with lower exhalation capacity. The high activities encountered in these materials may pose a threat to human health in case of using it for dwellings due to the gamma exposition. Nevertheless, with the goal of reducing the amount of waste generated in the mining industry, the application of the CMOC aggregate for concrete may be applied for the construction of outdoor structures such as bridges and viaducts.

Table 2: Exhalation rate for radon and thoron found in different construction materials

	^{222}Rn		^{220}Rn		Refer
Granite	0.06	0.01	76	34	[25]
	1.04	0.03	2420	667	
Granite	0.7		2515		[26]
	1.33		5900		
Cement	0.00049	0.00001			
Aerated concrete			75		[27]
Concrete			96		
Concrete	0.4	0.1			[28]
Sand	0.00029	0.00002			[29]
Sand	0.008	0.002			[28]
Aggregate	0.00150	0.00006			[28]
Aggregate	0.00044	0.00003			[29]
Brick	3.09		100		[26]
Brick	7.07		640		[26]

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